Large positive and negative lateral optical beam displacements due to surface plasmon resonance

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We report abnormally large positive and negative lateral optical beam shifts at a metal–air interface when the surface plasmon resonance of the metal is excited. The optimal thickness for minimal resonant reflection is identified as the critical thickness above which a negative beam displacement is observed. Experimental results show good agreement with theoretical predictions and the large observed bidirectional beam displacements also indicate the existence of forward and backward surface propagating waves at the surface plasmon resonance of the metal. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1775294]

As described by Newton,[1] when a finite size light beam is total internally reflected by a dielectric interface, the electromagnetic field partially penetrates into the rear medium and builds up an evanescent wave field whose amplitude decreases exponentially with the distance from the surface. The static Poynting vector of the evanescent wave is directed along the interface with a complex wave vector. Hence, after re-emerging into the former medium, the actual reflected energy flux is laterally displaced with respect to the geometrical optics beam. This effect, now known as the Goos–Hänchen (GH) effect, was experimentally demonstrated by Goos and Hänchen in l947.[2] Most investigations have examined this phenomenon by using a beam that is reflected from the interface of two dielectric media with the incidence angle sufficiently close to the total internal reflection (TIR) angle. Normally, the lateral displacement is proportional to the penetration depth which can be predicted by geometric optics[2,3] or analyzed by plane wave expansion models,[4,5] but the maximum longitudinal beam displacement is usually extremely small, because the penetration depth is of the same scale as the wavelength. However, large longitudinal beam shifts could be achieved by utilizing material[6,7] or structural resonances.[5,8] For example, two types of resonant multilayer structures were recently proposed by Schreier *et al.*,[9] which potentially could provide millimeter scale lateral shifts at optical wavelengths. Meanwhile, a negative lateral beam shift in reflection was discovered in the systems with absorptive materials,[10] negative refractive index media,[11] and/or resonant artificial structures.[12,13] As described by Tamir *et al.*,[5] such a negative displacement is due to the backward leaky wave with an opposite sign for the propagating and attenuation constant. Indeed, such a backward propagating surface wave can be easily excited on a metallic plasma slab with a negative permittivity under certain conditions.[14] Furthermore, the surface plasmon field amplifies the amplitude of this leaky wave through a resonance. Consequently, greatly enhanced negative beam displacements can be expected by carefully designing metallic surface plasmon resonant (SPR) structures.

In this letter, we present the theoretical and experimental examination of the beam displacement at visible wavelengths on a metal–air interface. As the SPR resonance is excited, we observe a lateral spatial displacement of greater than 50 wavelengths for the reflected beam due to the propagating surface wave. We also show that the greatly enhanced negative beam shift is observable when the film is thicker than a critical thickness, as determined by the dielectric properties of the noble metal. To excite the SPR, metal films (ε_1) with various thicknesses contact a high index medium (ε_0) as shown in Fig. 1, which is known as the Kretschmann–Raether attenuated total reflection (ATR) device.[15] Here ε_j are the permittivities of the different media of the prism, the metal layer and air (ε_2). At resonance, the incident electromagnetic field decreases exponentially in the film and excites the surface plasmon wave propagating along the metal–air interface. The exponential dependence of the evanescent electric field is schematically shown in the inset of Fig. 1. Generally, for transverse magnetic (TM) polarized incident light, the resonant electromagnetic field reaches its maximum at the interface and the typical enhancement factor is $\sim 10^2$ compared to the incident amplitude. On the contrary,

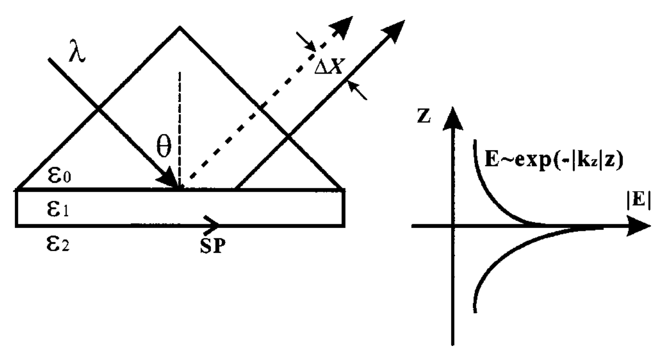

FIG. 1. Kretschmann–Reather ATR configuration, with the metal thin film (ε_1) coating. The solid reflective beam shows a positive optical beam shift compared to the geometrical reflection ray (the dashed line). The insets show the exponential dependence of the electrical field along the metal–air interface.

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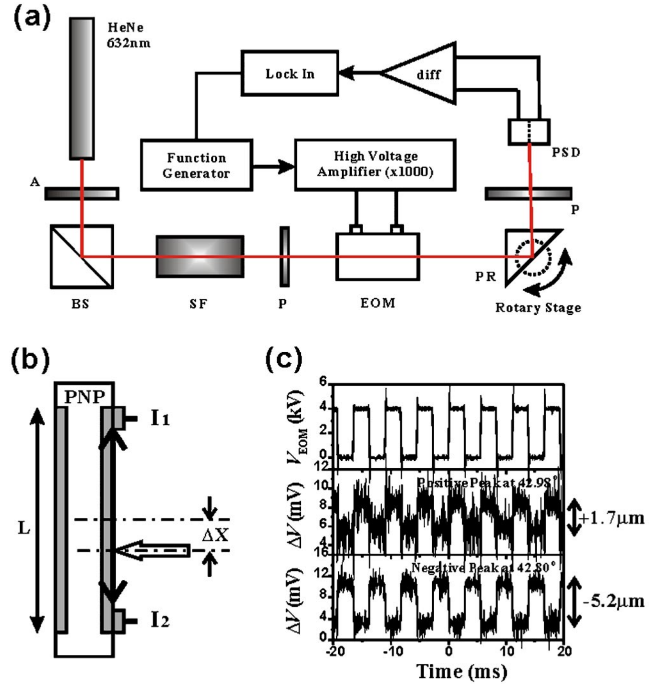

FIG. 2. (Color online) (a) Experimental measurement of the GH shift (A: Attenuator; BS: Polarized beam splitter; SF: Spatial filter; P: Polarizer; EOM: Electro-optical modulator; PR: Prism with metal coating; and PSD: Position sensitive device). (b) Schematic view of a PSD device producing two photocurrents which are proportional to the incident beam position. (c) External modulation voltage (panel 1), and differential voltage signals from the PSD device indicating the positive (panel 2), and negative (panel 3) lateral beam displacement. The positive ($+1.7\ \mu$m) and negative ($-5.2\ \mu$m) displacements are measured at the incident angles of 42.98° and 42.80° on a 82.0 nm silver film.

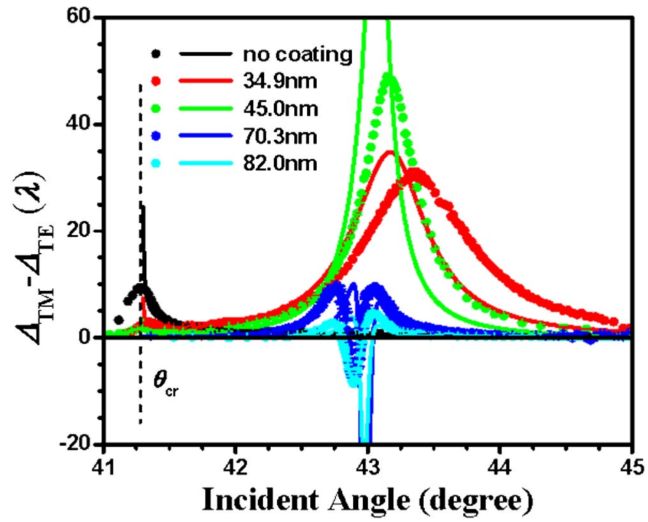

FIG. 3. (Color online) Measurements of relative lateral beam shifts on silver films with various thicknesses. The calculated relative displacements are plotted for comparison as solid curves.

there is no enhanced plasmon field with transverse electric (TE) excitation.

Next, we consider a finite size optical beam impinging upon the above described ATR device. Due to the enhanced electromagnetic field at the metal–air interface, intuitively and substantiated by measurement and calculation, much more energy penetrates into the rear medium as compared to the normal TIR case without the metal layer. Therefore, the enhanced surface wave effectively enlarges the lateral beam displacement in the SPR case. Furthermore, if a backward propagating surface wave exists, the lateral displacement should be negative with respect to the geometrical optics beam, where the time-averaged Poynting vector is directed along the interface and pointed in the opposite direction of the incident wave vector.

However, direct experimental measurement of such a beam shift from a single reflection has always been difficult in the past due to the small value of the displacement and the diffractive property of the laser beam.[16] In this letter, a highly sensitive modulation detection scheme was applied following the original experiment of Gilles *et al.*[17] by combining an electro-optical polarization modulator (EOM) and a one-dimensional position-sensitive detector (PSD). As schematically shown in Fig. 2(b), a PSD device provides two signals that are proportional to the barycentric position of the incident beam. The lateral displacement of the impinging beam is hence proportional to the normalized differential output of the detector.

The schematic diagram of the experimental setup is shown in Fig. 2(a). The surface plasmon excitation is achieved by impinging a HeNe laser beam (632.8 nm), with a waist of about 200 μm, onto a thin silver film which is coated on a right angle BK7 prism. The silver film is coated by electron-beam deposition with various thicknesses and the incident angle is controlled between 40° to 45° by a motorized stage, which covers both the TIR angle (41.3°) and SPR angle (about 42.8°) for the above ATR device. The polarization states of the incident beam are controlled by an EOM (PKC21, Inrad Inc.), which is driven by a square wave provided by a high-voltage amplifier (Trek610, Trek, Inc.). The half-wave voltage is $\sim$4.2 kV and the typical modulation frequency is 200 Hz. The modulation depth (extinction ratio) is larger than 100:1 in the experiment. The reflected beam from the prism passes through a polarized analyzer and is collected by a PSD detector (S7879, Hamamatsu, Inc.) and a lock-in amplifier measures the spatial displacement. By periodically modulating the incident polarization, the difference of the lateral displacement between TE and TM light is measured. However, since the TE polarized incidence cannot excite any surface plasmon polariton, there is no enhanced lateral displacement. Therefore, it serves as a perfect reference beam and the measured relative beam shift between TM and TE excitation indeed indicates an absolute beam displacement for a TM wave at the SPR region. One set of detected positive and negative beam shift signals is plotted in Fig. 2(c) for comparison. Both the positive (panel 2) and the negative (panel 3) displacement signals are compared to the external voltage (panel 1) applied onto the EOM, which controls the incident polarization states. Unlike the positive shift signal, the negative displacement signal is out of phase with the driving voltage. In this case, although the measured negative shift is the relative displacement between the TM and TE wave, the TM polarized reflective beam undergoes an absolute negative shift due to the absence of a surface plasmon resonance for the TE polarized illumination.

With the above convention, the measured relative beam shifts ($\Delta_{TM}-\Delta_{TE}$) for various thicknesses of silver films are plotted in Fig. 3 respective to the incident angles. The bare BK7 glass prism without a silver coating is also measured and plotted as a black curve for comparison, which is fitted by the Artmann equation[3] (shown as black solid curve in Fig.

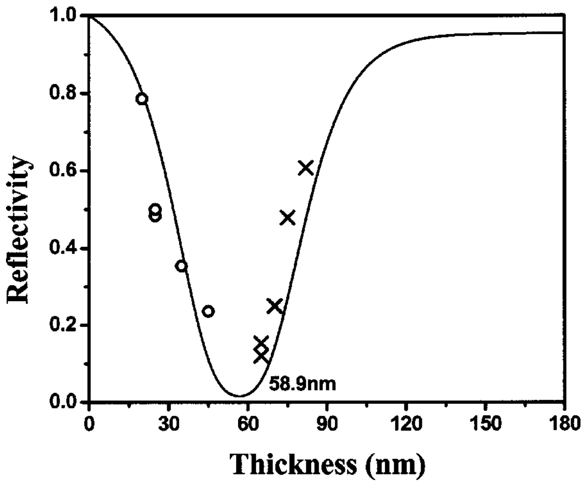

FIG. 4. Thickness dependence of the reflectivity for TM polarized excitation is shown by the solid curve. The experimental results are plotted as separate points (○: Positive shifts only; ×: Negative shifts observed) and the minimum indicates the critical thickness.

3) and describes the GH shift at the critical angle of the dielectric interface. The enhancement of the lateral displacements is observed at the SPR angle on all the silver coated prisms. For thinner silver films less than about 60 nm, as the thickness increases, the maximum relative displacement increases significantly. A shift of as high as 50 wavelengths is observed on a 45 nm silver film. Additionally, the relative lateral beam displacement at the resonant angle becomes negative for films thicker than about 60 nm, as shown in Fig. 3.

To understand such interesting behavior, we studied the lateral displacements using a stationary-phase analysis.[3] Quantitatively, such a shift is determined by the phase retardation of the reflected wave, $\Delta = -\lambda / 2\pi\sqrt{\varepsilon_0}\, d\Phi / d\theta$, where λ is the incident wavelength, θ is the incidence angle, and Φ is the phase difference between the reflected and incident waves. By assuming a wavelength of 633 nm with dielectric constants of $\varepsilon_0 = 2.23$ (BK7), $\varepsilon_1 \approx -18 + 0.5j$ (silver at 633 nm),[18] and $\varepsilon_2 = 1$ (air), the angular dependence of the beam shifts at the metal–air interface with both TE and TM polarized excitation are calculated. Due to the absence of the resonance, in the simulation, no significant beam displacement (larger than one wavelength) was observed for TE polarized illumination except in the vicinity of the critical angle. However, for TM polarization, greatly enhanced lateral displacements are observed at the SPR angle. Moreover, negative displacements were also observed as the thickness of the metal films exceeded a certain critical value, which indicates the excitation of the backward propagating surface wave due to the extremely large spatial dispersive behavior of the retarded phase at the plasmon resonance.[19] For comparison with the experimental results, the relative displacements for the same silver ATR devices are plotted in Fig. 3 as solid curves, which show good agreement with the experimental results. The expected displacement peak is typically higher and has a narrower angular tolerance compared to the experimental results, which is most probably caused by additional scattering due to the surface roughness of the deposited film.[15]

In Fig. 4, the thickness dependence of the reflectivity for TM polarized excitation is shown as a solid curve. In Fig. 4, the observed data points for both positive and negative lateral beam shifts are plotted as separate points. As shown in the figure, the thickness for a reflection minimum indicates the critical condition at which the negative beam displacement appears. This thickness is also well known as the optimal thickness for excitation of the surface plasmon,[20] where the resonance absorption is just balanced by radiation damping and internal damping. By assuming $|\varepsilon_1'| \gg \varepsilon_1''$, ε_0, ε_2, which is normally true for the above described ATR system at visible wavelengths, the minimum of the reflectivity determines the critical thickness,

$$d_{\mathrm{cr}} = \frac{\lambda}{4\pi\kappa} \ln \frac{2\kappa}{n},$$

which predicts that the critical thickness in this experiment should be 59 nm as indicated in Fig. 4. Here, n and κ are the refractive index of the metal $(n + j\kappa = \sqrt{\varepsilon_1})$. Furthermore, by comparing Figs. 3 and 4, the much larger longitudinal displacement can be expected as the thickness approaches the critical thickness.

In conclusion, a large positive and negative lateral beam displacement is observed on the silver–air interface when the surface plasmon resonance is properly excited. The condition for a negative beam shift is predicted theoretically and confirmed experimentally. Such positive and negative spatial displacements indicate the existence of forward and backward surface propagating energy flows along the metal–air interface at resonance. Furthermore, the measurement of beam shifts provides an alternative SPR sensing scheme, which potentially improves the sensitivity of SPR sensors significantly.

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